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A review of biomass burning emissions part III: intensive optical properties of biomass burning particles

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Abstract. Because of its wide coverage over much of the globe, biomass burning has been widely studied in the context of direct radiative forcing. Such study is warranted as smoke particles scatter and at times absorb solar radiation efficiently. Further, as much of what is known about smoke transport and impacts is based on remote sensing measurements, the optical properties of smoke particles have far reaching effects into numerous aspects of biomass burning studies. Global estimates of direct forcing have been widely varying, ranging from near zero to $-1 \,\mathrm{W} \,\mathrm{m}^{-2}$. A significant part of this difference can be traced to varying assumptions on the optical properties of smoke. This manuscript is the third part of four examining biomass-burning emissions. Here we review and discuss the literature concerning measurement and modeling of optical properties of biomassburning particles. These include available data from published sensitivity studies, field campaigns, and inversions from the Aerosol Robotic Network (AERONET) of Sun photometer sites. As a whole, optical properties reported in the literature are varied, reflecting both the dynamic nature of fires, variations in smoke aging processes and differences in measurement technique. We find that forward modeling or "internal closure" studies ultimately are of little help in resolving outstanding measurement issues due to the high degree of degeneracy in solutions when using "reasonable" input parameters. This is particularly notable with respect to index of refraction and the treatment of black carbon. Consequently, previous claims of column closure may in fact be more ambiguous. Differences between in situ and retrieved ω_o values have implications for estimates of mass scattering and mass absorption efficiencies. In this manuscript we

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review and discuss this community dataset. Strengths and lapses are pointed out, future research topics are prioritized, and best estimates and uncertainties of key smoke particle parameters are provided.

1 Introduction

Before the effects of smoke particles on the earth's radiative balance can be known, their optical properties need to be efficiently parameterized. Further, these parameterizations need to be physically consistent with the particle's other physical and emissions properties, which can vary significantly from region to region. Fundamental input parameters such as index of refraction and black carbon content are highly uncertain. Because smoke particles size range is in the steepest part of the scattering versus physical cross section curve, even small changes in estimated physical parameters can have significant impacts on scattering and absorption efficiencies. The result is a considerable amount of degeneracy in "closure" calculations and relatively easy justification for any experimental or modeling finding based on "physically sound" assumptions or parameterizations. This review paper is concerned specifically with these issues.

This review paper is the third of four examining biomassburning emissions and relies heavily on Part II (Reid et al., 2004), which deals with particle size and chemistry issues. Here we evaluate the radiative impacts of smoke particles along three principle lines: 1) Bulk parameterization from measurement, 2) direct forward calculation based on particle size distribution and chemistry, 3) the inverse problem where flux and radiance values are related to an optical equivalent size distribution. We begin with a review of field

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measurements of key optical parameters. We then compare these findings with forward modeled studies and column closure experiments. These are subsequently compared to solutions from inversion methods. In all of these sections we explore differences in particle properties by region and fire chemistry, and attempt to reconcile differences that exist between investigation techniques. In conclusion we discuss our findings and present what we feel are reasonable parameters with likely uncertainties for smoke properties. Suggestions are made for future research.

6 Conclusions

In this manuscript, we provide a short review of the optical properties of biomass burning particles. Estimates from in situ measurements, forward calculations, and inversions studies are compared. In the end, we give best estimates for median values of smoke optical properties, knowing full well that each fire has its own character and can deviate significantly from the mean. The main points of the review are summarized below.

- Over the past two decades, measurements of particle mass scattering (α_s) and absorption (α_a) efficiency have been relatively consistent. As these properties are strongly correlated to particle size and black carbon content, their variability is strongly tied to individual fire physics. For example, flaming combustion produces smaller α_s and larger α_a and ω_o compared to smoldering combustion. Consequently, optical properties of fires change rapidly as they go through their lifecycles.
- Just as aging processes affect smoke particle size and chemistry, they have a significant influence on smoke particle optical properties. Measurements of particle properties made near fires are difficult to apply to large regional smoky-hazes. Coagulation keeps particle black carbon ratios constant, but will resulting an increases α_s and ω_o due to the increase in size alone. Condensation or out-gassing processes will increase α_s and ω_o and reduce α_a .
- Smoke particle hygroscopicity is uncertain, with the only two direct measurements in the literature yielding different results. Values derived from inversion methods yield an even larger spread. Almost no data have been presented on particle hysteresis effects.
- We show that there is a wide divergence in forward modeling or "internal closure" calculation methodologies, with differences based in unconstrained assumptions on density, size, black carbon content and index of refraction throughout the literature. While such calculations can be gratifying, ultimately the high degree of freedom in input parameters makes such studies less useful than as typically presented.

- Particle index of refraction is highly uncertain, and is often treated as a free parameter. Differences in the literature can alter the computed α_s , α_a , and ω_o considerably.
- Early inversions studies show very inconsistent results with derived values that were unphysical. Recent inversion studies are better constrained, and show consistency with what is qualitatively known about various biomass-burning regions of the world. However, as in forward modeling, there is a possibility of degeneracy in the solutions. This may lead to variability in estimated values of the mass scattering and absorption efficiencies.
- While it has been argued that the bulk of in situ particle measurements overestimate absorption, the bulk of measurements near sources have been made by extinction cell (hence this argument does not hold as well). However, such arguments have merit for regional smoke. Even so, after corrections are made, derived α_a values are still considerably higher than what is given by inversions.
- Lastly, we provide estimates of fine mode smoke particle properties. For the most part, these are consistent with what was suggested by IPCC (2001), although we suggest a higher mass scattering efficiency and hygroscopic growth factor.